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## **Recent Developments in Neutron Detection and Multiplicity Counting with Liquid Scintillator**

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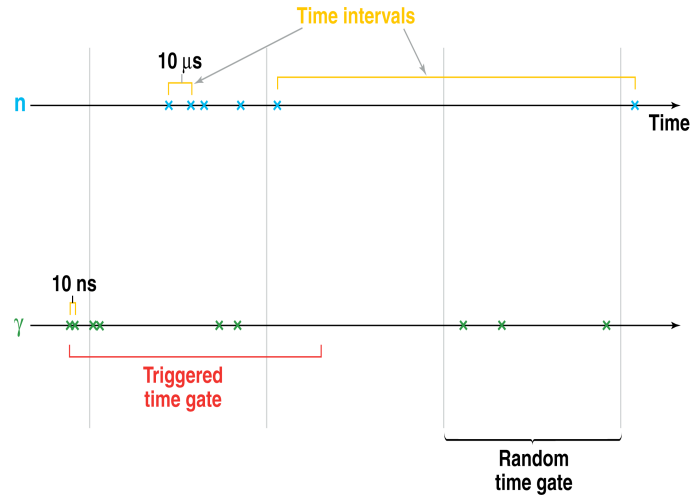
### **0. Abstract**

For many years at LLNL we have been developing time-correlated neutron detection techniques and algorithms for many applications including Arms Control, Threat Detection and Nuclear Material Assaying. Many of our techniques have been developed specifically for relatively low efficiency (a few %) inherent in the man-portable systems. Historically we used thermal neutron detectors (mainly  $^3\text{He}$ ) taking advantage of the high thermal neutron interaction cross-sections but more recently we have been investigating fast neutron detection with liquid scintillators and inorganic crystals. We have discovered considerable detection advantages with fast neutron detection as the inherent nano-second production time-scales of fission and neutron induced fission are preserved instead of being lost in neutron thermalization required for thermal neutron detectors. We are now applying fast neutron technology (new fast and portable digital electronics as well as new faster and less hazardous scintillator formulations) to the safeguards regime and faster detector response times and neutron momentum sensitivity show promise in measuring, differentiating and assaying samples that have very high count rates as well as mixed fission sources (e.g. Cm and Pu). We report on measured results with our existing liquid scintillator array and progress on design of nuclear material assaying system that incorporates fast neutron detection.

### **1. Introduction**

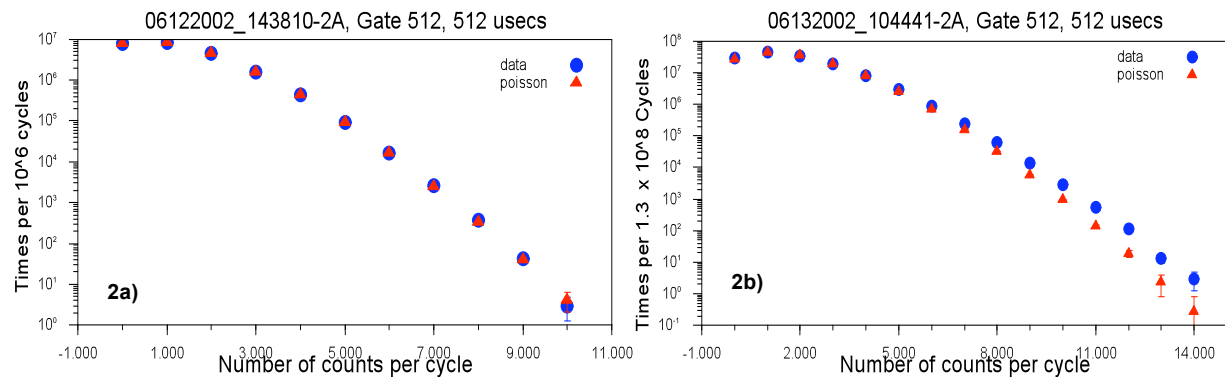
The low natural background rates and the penetrating nature of neutron radiation makes neutron detection a good method for quantifying and accounting for large amounts of special nuclear material capable of neutron induced fission and fission chains. Fission is one of the few natural processes that produces time-correlated neutrons, the others are spallation-type processes (e.g. (n,xn), cosmic induced background, etc) which have low but measurable rates in common terrestrial material. The high rates of most transuranic spontaneous fission sources of even a gram or less usually swamp any cosmic induced background rate but even kilogram quantities of natural uranium produce neutrons only on the same order as that of the cosmic generated background flux. However the primary characteristic of special nuclear material is its ability to fission and to support fission chains through neutron induced fission. This means that neutrons produced from fission are not produced randomly singly but rather in time-correlated bursts. Almost from the beginning of the atomic age, the measurement of time-correlated neutrons has been used to detect and quantify fission and fission processes. Feynman himself proposed a method, the grandfather of what is used today, to compare correlation rates of neutron flux in a fixed time to that which would be expected of a random neutron source.

## 2. Measurements with $^3\text{He}$ Thermal Neutron Detectors



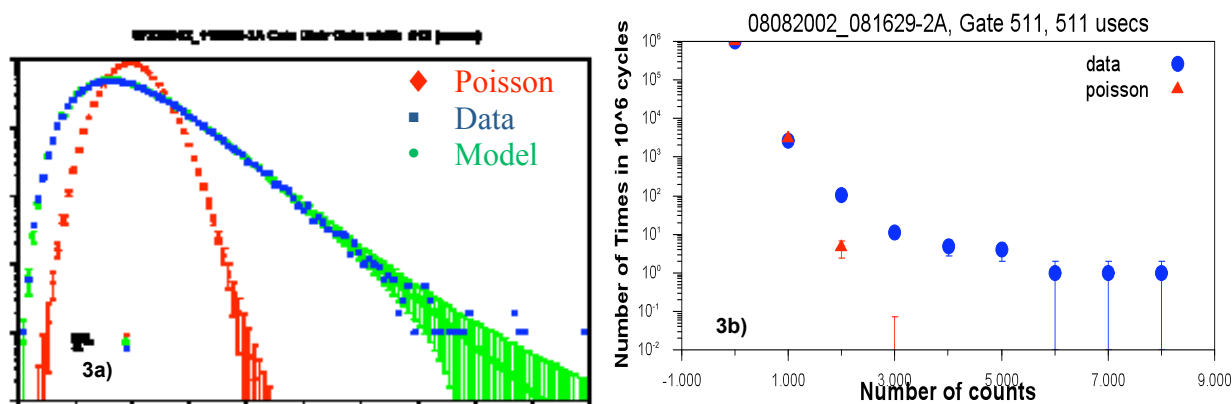
**Figure 1. Three different ways to measure a time series 1) The time between arrivals 2) Counts in a fixed time gate triggered by an arrival and 3) Counts in a randomly triggered fixed time gate (all data shown here)**

There are many ways to measure time correlations, three examples are illustrated in Figure 1). One can measure the time intervals between arrivals, one can measure a fixed time interval from a detected neutron or one can count the number of neutrons occurring in a randomly triggered event. We have a long history of developing such methods based on counting detected neutrons in randomly triggered time-gates. Figures 2 and 3 are data from portable  $^3\text{He}$  thermal neutron detectors that can detect and assay



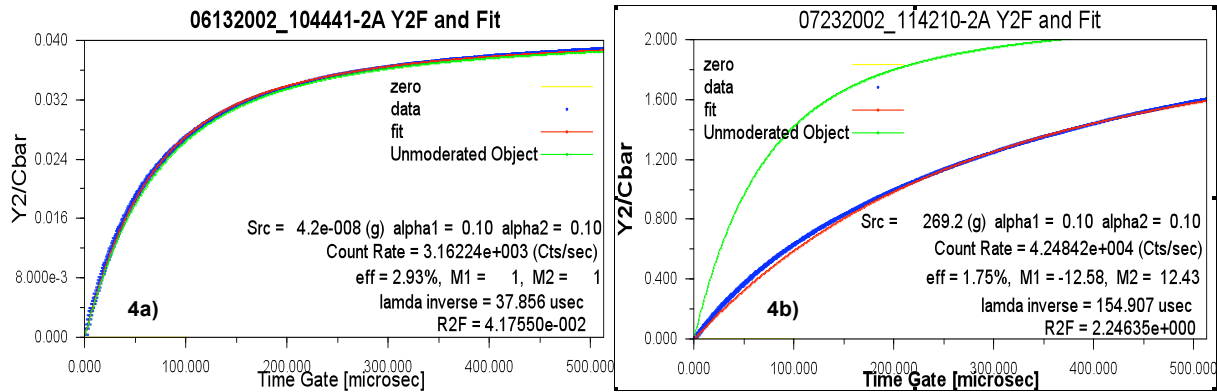
**Figure 2. Data from two sources 2a) an AmBe source (2.1K cts/s) and 2b) a  $^{252}\text{Cf}$  source (3.1K cts/s 3% efficiency). The Data are the number of neutrons detected (x axis) in 0.512 ms vs. log number of times (Y axis). Wider Variance of data from Poisson distribution of same count rate is indication of Fission fissioning sources even though it is only a 1-3% efficient.**

The data shown are from a single time gate of 512 microseconds. In practice we vary the time gate from 1-512 microseconds and compare all the data to a theoretical model to find a best solution. Figure 2) shows how comparing a measured count distribution from a random (AmBe -alpha-n) source can be easily visually distinguished from a spontaneous fission source ( $^{252}\text{Cf}$ ) simply by comparing the measured distribution (blue) from that expected from a random source of the same count rate (red). In the case of the fissioning  $^{252}\text{Cf}$  source the variance of the distribution is significantly wider than that expected if the source were purely random. This difference is accentuated in the case of multiplying objects such as a highly multiplying plutonium ball shown in Figure 3a). This also works even for weak sources like the modestly multiplying uranium shown in Figure 3b). In this case the actual count rate is very low (only a couple net counts/second on a background of 4 counts/second) but those few originating in the uranium are highly correlated.



**Figure 3. Data from 3a) a strong source, plutonium ball (52s data, 42.5K cts/s, M=12.5, efficiency 2%) source (2.1K cts/s) and 3b) a weak source uranium shell (500s data, 2.1 net cts/s M=2.5). Data is still highly correlated.**

In any case, methods that we have been perfecting also allow assaying of the material. We do this by predicting the distributions ( e.g. in the above cases from 1-512 microseconds) we would see given a source-strength, a multiplication, an alpha ratio, a detection efficiency and a time of correlation (for bare objects measured with a thermal neutron detector this will be the detector thermalization time) and finding the best fit of the data to our predictions. In cases where only the neutron source may be measured directly, the neutron source strength can be then combined with material isotopics gathered from other means (such as gamma ray spectroscopy) to determine a total mass. The data required for a few % assay can be obtained in a minute or less for strong significantly multiplying sources (like the plutonium source shown in Figure 8) to perhaps a hour or more in the case of a weak source (like uranium) or a completely non-multiplying source like  $^{252}\text{Cf}$ .

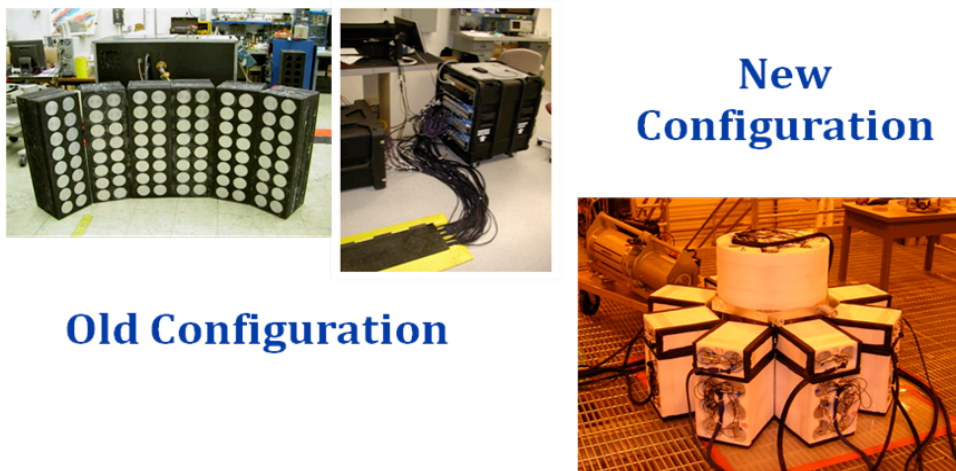


**Figure 4. Second Moment Y2 vs Gate width 1-512 microseconds for 4a) unmoderated  $^{252}\text{Cf}$  and 4b) heavily moderated plutonium ball.**

We also find it useful to plot the moments as a function of the varying time gate width. The evolution of the normalized moments vs. the gate width can give indications of moderation which is illustrated in Figure 4) with data from an unmoderated  $^{252}\text{Cf}$  source and a heavily moderated plutonium ball. The green curve is a prediction for the evolution with the known detector time constant ( $\sim 40$  microseconds). The match with the  $^{252}\text{Cf}$  data in 4a) confirms little or no moderation and the much slower approach to its asymptotic value is an indication of heavy moderation in 4b).

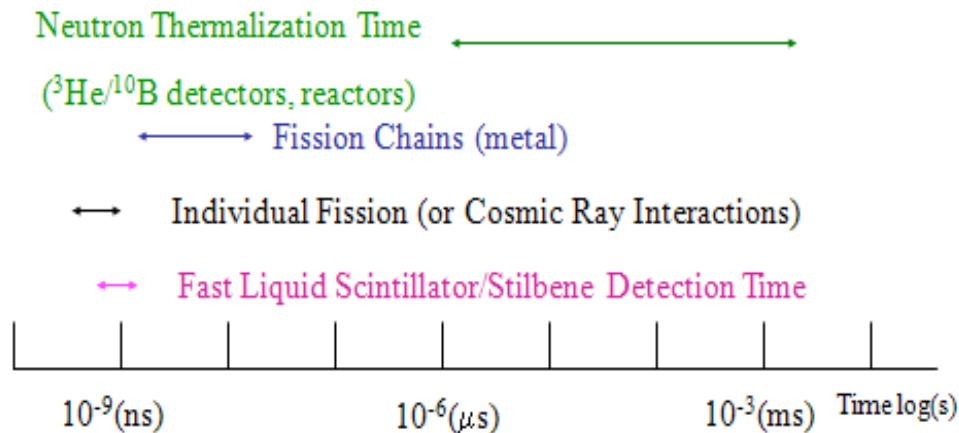
### 3. Measurements with Fast Liquid Scintillation Detector Array

Recently we have begun to apply our neutron analysis and assaying techniques to fast neutron detection with liquid scintillators. In Figure 11 we have configured an array of liquid scintillators so they physically cover about  $2\pi$  of the solid angle off a cylindrical chamber at the center of the array. Fast neutron detection relies on the recoil of a charged particle (most likely a proton) from collision with a neutron. The clear disadvantages of fast neutron detection are 1) the lower neutron scattering cross-sections (on the order of a barn at best) 2) the minimal neutron energy required for a recoiling proton to be seen (around 1 MeV) and 3) the added difficulty of accurately identifying a neutron recoil proton from a gamma ray. Fast neutron detectors can not possibly be as efficient as the most efficient thermal detectors simply because of the minimum energy detection threshold for fast neutron detectors and correspondingly they can not be as efficient per unit weight. Efficiency is of course a very important factor when considering measuring correlated events as the probability of detecting  $n$  correlated neutrons goes as the  $n$ th power of efficiency.



**Figure 5. Liquid Scintillator Arrays 5a) Old configuration and 5b) more efficient new configuration.**

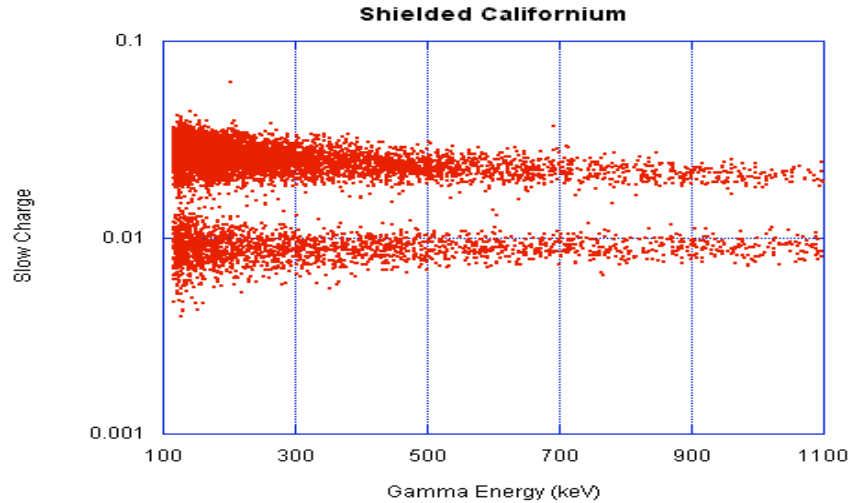
However there are several very important advantages to fast neutron detection which can be paramount, especially in areas where thermal neutron detection has broken down as in high flux. In high fluxes the probability of random correlations increases geometrically which makes the ability to detect fission correlations increasingly difficult. Consider the measurement of  $^{252}\text{Cf}$  in the Figure 2b).  $^{252}\text{Cf}$  only rarely fissions with a neutron multiplicity greater than 8 and yet with a detector with 3% efficiency the detector saw a non-negligible amount of times when more than 10 neutrons were detected in the detector. This clearly means that with a modest source flux of a 100,000 neutrons/second (detecting 3,000 neutrons/second) there is a significant amount of overlapping of fission events. The single most important characteristic of fast neutron detection is that it happens fast. Fast neutron detection allows the relevant detection time to shrink from ten's of microseconds (detector thermalization time) to nanoseconds which is equivalent to reducing the effective flux by a factor of 10,000.



**Figure 6. Illustration of timescales for Fission and Fission chains both occurs at a much shorter time scale than thermal neutron detector can detect. Fast Liquid and crystal scintillators are fast enough to discriminate time scales of individual fission from fission chains**

Secondly the fast detection preserves the timescale of the original neutron production. The prompt production of fission neutrons (from a single fission) and spallation-type processes occurs on a nanosecond timescale while the neutron production of a multiplying body occurs in the neutron transit time which can be 10's of nanoseconds for pure metallic systems and up to many microseconds for moderated systems (Figure 6). Thermal neutron detection (e.g.  $^3\text{He}$ ,  $\text{BF}_3$  etc.) requires moderation of the neutrons for efficient detection and this occurs on a ten's of microsecond timescale which loses all the timescale details of the original neutron production.

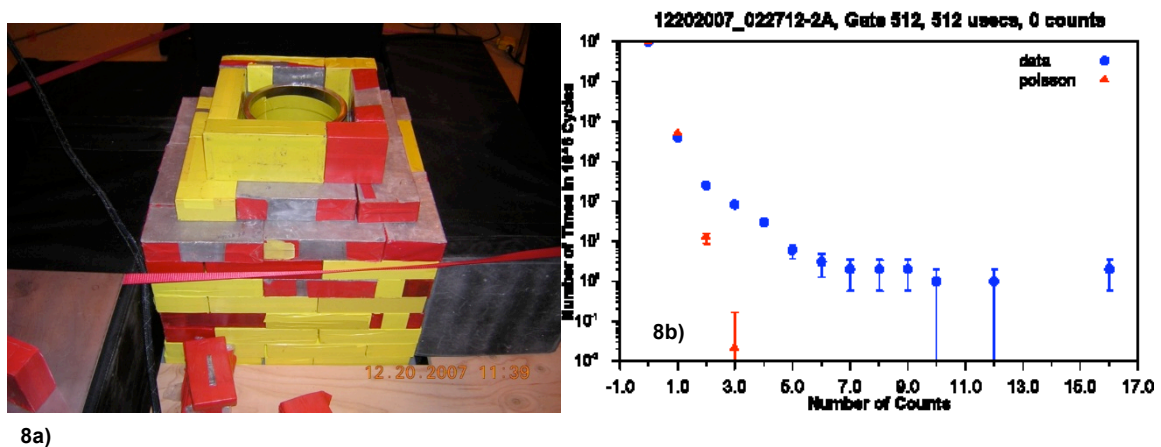
Finally fast neutron detection also preserves some of the original neutron energy spectral information since it works on recoil energy. Nearly all neutrons detected by thermal detection are by definition thermal neutrons so almost all initial energy information is ultimately lost. Also more subtly, especially when combined with the introduction of low energy (below detection threshold) neutrons measuring the *change* in neutron flux can be most illuminating with respect to the source of neutrons in a sample.



**Figure 7. Clean separation of gamma rays (top) from neutrons (bottom) employing digital electronic discrimination.**

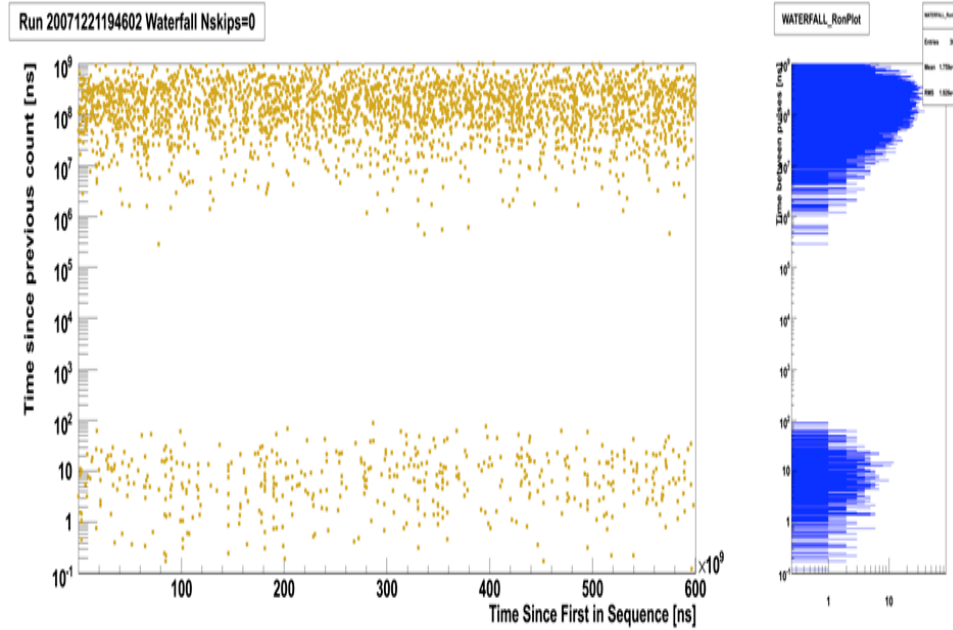
Figure 7 illustrates the now very clean ability to discriminate neutron events from gamma rays due to the advent of modern fast digital electronics. Photon rejection can be done nearly perfectly if full digitization of a pulse is employed as the normal confusion of a later arriving gamma ray can actually be seen and rejected.

The last figures are meant to illustrate the power of a fast neutron detection system with data taken with the existing liquid scintillator array. In Figure 8) an approximately 1 ton pile of lead bricks was measured with thermal neutron detector's (approximately 4% efficient). Very large neutron correlations were seen at the 10's of microsecond time scale. Compare this to Figure 3b) of a multiplying uranium system and one would be very hard pressed to tell the difference. In fact assuming the pile of lead were a multiplying uranium system one could get neutron distribution which looked quite close.



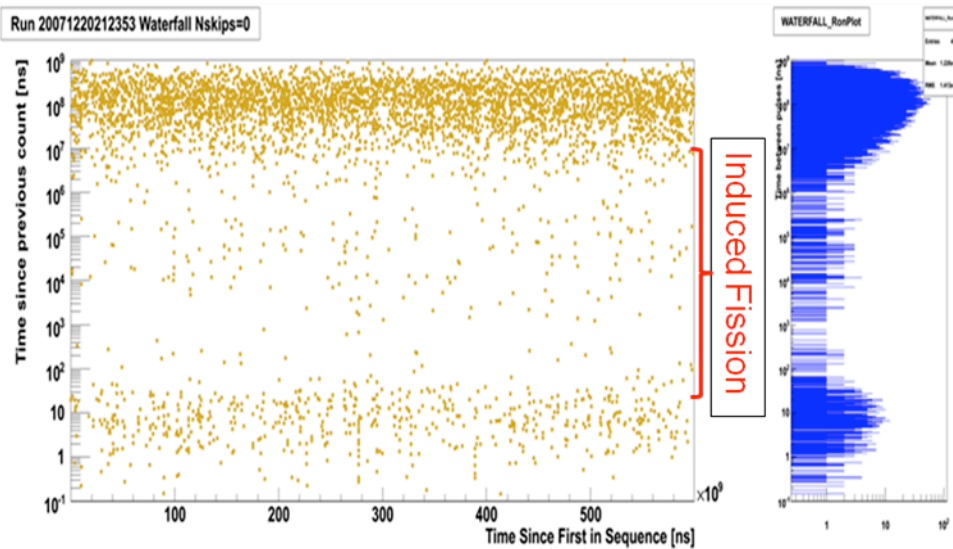
**Figure 8. Thermal Neutron Data taken on a ~1 Ton Pile of Lead (Compare to plot 3b). Scale which is the average count rate ( 6 counts /second). The lower band is fast time correlations which are generated by cosmic interactions with the lead pile**





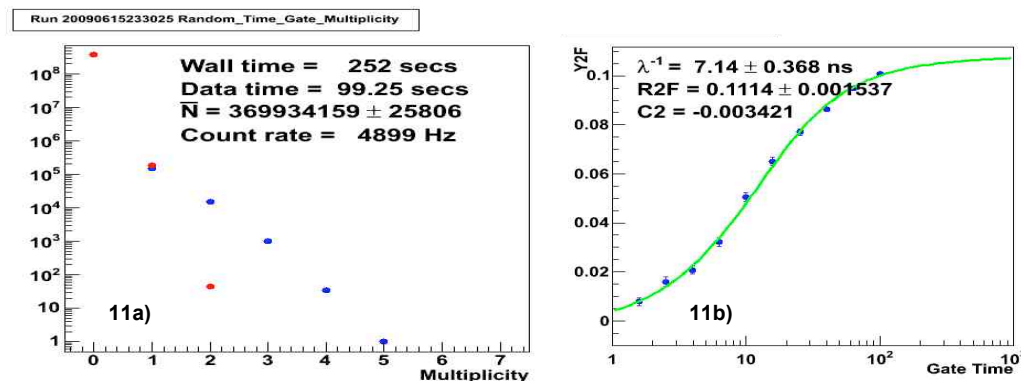
**Figure 9. Fast Liquid Scintillator Neutron Data taken on ~1 Ton Pile of Lead (old configuration). 600s of data plotted neutron arrival times (x axis) vs. log (time) to next arrival (Y axis). Note the absence of correlations in the 40ns to 1 microsecond time scale.**

We measured the pile of lead with our 1-2% efficient liquid scintillator array (old configuration in Figure 6) in Figure 9. Here the y-axis is the log of the time interval between neutron counts (in nanoseconds and each tick is a power of nanoseconds) and the x-axis is linear running time (for 600 seconds). What you see is the top wide band which happens on the 10 microsecond to second time scale which is the average count rate ( 6 counts /second). The lower band is fast time correlations which are generated by cosmic interactions with the lead pile. They occur and are over in less than 10 or 20 nanoseconds. There are no time correlations occurring in the time scale between a few ten's of nanoseconds and 1 microsecond.



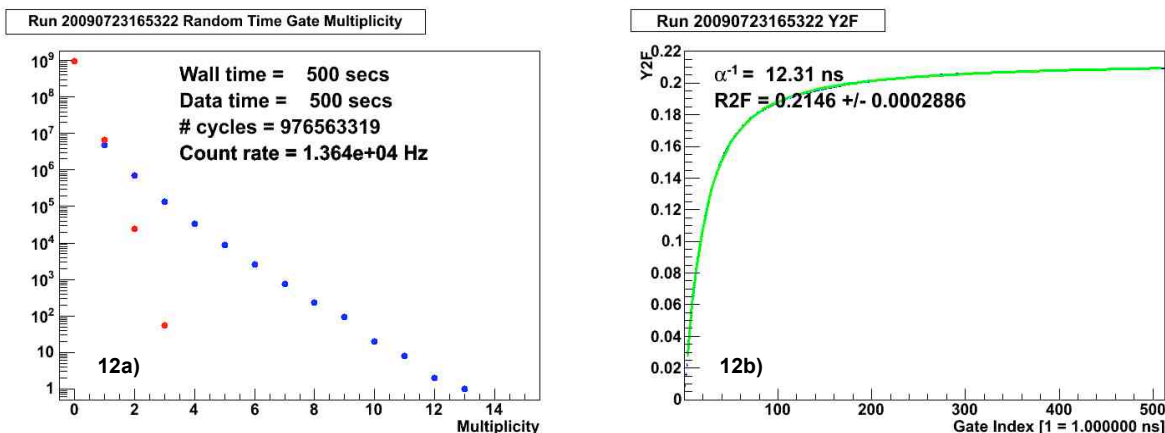
**Figure 10. Fast Liquid Scintillator Neutron Data taken on ~1 Ton Pile of Lead (old configuration) plus embedded multiplying Uranium. Note the correlations in the 40 ns to 1 microsecond time scale this is a clear indication of the presence of induced fission.**

The Figure 10) is the same pile of lead with some multiplying uranium embedded inside. You would see no gamma ray signatures and the thermal detector would still look correlated but a lot like the data in Figure 8) but the measurement reveals time correlations occurring in the intermediate time scale where nothing occurred before. This is a clear indication of the presence of nuclear material as this intermediate time scale can only occur because of slower neutrons inducing fission in the uranium which can not happen in lead alone.



**Figure 11. Fast liquid scintillator neutron data taken on  $^{252}\text{Cf}$  source 99.25 s data with a 511 ns gate (4.9K cts/s ~6% efficiency). Note the clean separation from Poisson distribution with same count rate because of short time gate possible from fast timing.**

The last two figures are demonstrations of assaying done with the liquid scintillators on real objects and the advantage of fast timing in reducing the random correlations. In Figure 11 a  $^{252}\text{Cf}$  source was measured in the new configuration liquid scintillator array that is about 6% efficient. Even with a higher count rate than was measured in the thermal neutron measurement of Figure 2b), there is clearly very little random correlations and with 4 minutes of real time (100 seconds of data) as assay was easily accomplished compared to the 8.5 hours of data shown in Figure 2b).



**Figure 12. Fast liquid scintillator neutron data taken on plutonium source 500s data with a 511 ns gate (13.4K cts/s ~5% efficiency). Note the clean separation from Poisson distribution with same count rate because of short time gate possible from fast timing.**

This also extends to plutonium systems. A small multiplying plutonium ball was measured in Figure 12 and an assay good to a few % accuracy was completed with 5 minutes of data. It is also important to note that the time constants seen in Figure 11b) of 7ns in the  $^{252}\text{Cf}$  source (a non-multiplying fission source) and the 12ns seen in Figure 12b) for the metal plutonium source. This implies that the measured timescales of a system may be the most significant detectable difference between a system with significant material able to support induced fission from slower neutrons and those without.

#### **4. Summary and Conclusions**

We have taken fast neutron data and performed assays with our liquid scintillation array good to a few % with a few minutes of data and been able to achieve 5 to 6% total efficiency with an approximately  $2\pi$  solid angle detector by applying algorithms developed originally for low efficiency portable thermal detectors. The intrinsically fast detection time of the liquid scintillator arrays greatly reduce random time-correlations inherent in time-correlation measurements and also permits direct measurement of the time scale differences between individual fission or fission-like processes (such as cosmic induced background) and those of fission chains which are unique to nuclear material able to support induced fission from lower energy neutrons. We believe that it is possible through direct measurement to differentiate between complicated samples with different fractions of spontaneous fission sources for example Cm vs. Pu or Cf which all have different averages of neutrons produced. We also believe that there is a wealth of information in the detailed timing information that can help with MC&A especially in samples with high fluxes and complicated neutron sources.. Not to mention the potential neutron energy information available to fast neutron detection that is not available to thermal detectors.

The inherently lower cross-sections for fast neutron detection will probably prevent fast neutron detection from replacing thermal neutron measurements in all cases but gas scintillator detection shows great promise for attacking some of the more difficult problems in the safeguards arena such as dealing with high flux from spent fuels for accountability or to deal with the possibility of nuclear material diversion. Particularly with the advent of possible new crystal scintillators which can replace the flammable liquid scintillators.

#### **Acknowledgements**

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